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Title: Ultrafast Investigation of Shock Induced Changes in GAP and Nitrocellulose Thin Films

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Ultrafast Shock Induced Changes in Glycidyl Azide Polymer and Nitrocellulose Thin Films

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Abstract:

Planar shocks generated in Aluminum(Al) by 120 femtosecond laser direct drive are impinged upon thin films of glycidyl azide polymer (GAP) and nitrocellulose (NC). The shock breakout in these thin film multilayers is monitored in real time with femtosecond spatial interferometry. The initial particle velocities were 0.9 km/s in GAP, and 0.5 km/s in NC, compared to 0.6 km/s for the Al drive layer. The phase change in the interferometric results at times greater than 20 ps exhibited odd behavior suggesting some combination of rarefaction waves arriving at the interface, reaction pushing back on the interface, and/or optical changes in absorption and refractive index of the polymer occurring upon shock. Further experiments are described to ascertain the influence of each possibility and to more fully characterize the shocked energetic and possible reaction.

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Los Alamos National Laboratory

High Explosive Reaction Chemistry via Ultrafast Laser Excited Spectroscopies

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Purpose

- Characterize flow of energy from a shocked lattice into individual energetic molecules
  - Multiphonon up-pumping? Internal conversion following direct electronic excitation upon compression?
- Determine initial reaction mechanisms and kinetics
  - How does reaction follow the shock in these high temperature, high pressure, nonequilibrium environments?
- Provide molecular level picture to aid in accurate scaling of predictions to larger, real world problems
  - How does molecular structure and condensed phase environment determine factors such as sensitivity?

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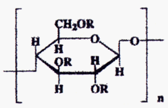
Energetic Polymer Thin Films

Nitrocellulose (NC)

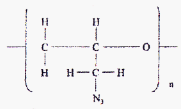
- Exothermic reaction at ~190^{0}C releases NO_2 gas



R: NO_2 or H

Glycidyl Azide Polymer (GAP)

- Exothermic reaction at ~ 240^{0}C releases N_2 gas



- Spin coated films from 50 nm to several microns thick
- Surface roughness <10 nm over 80x80 μm area (AFM)
- Both reactions will exhibit clear spectroscopic changes

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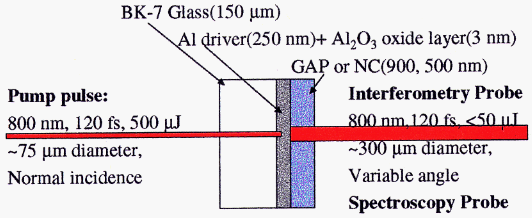
Experimental Details

Side View of Sample



Pump pulse:
800 nm, 120 fs, 500 μJ
~75 μm diameter,
Normal incidence

Interferometry Probe
800 nm, 120 fs, <50 μJ
~300 μm diameter,
Variable angle

Spectroscopy Probe
2nd pulse tunable
300 nm - 10 μm

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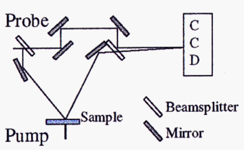
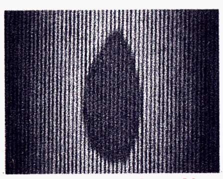
Spatial Interferometry

- Probe split, half reflects off Al through GAP or NC overlayer
- Pulses recombined in CCD
- Time resolved 2D spatial picture of surface with shock breakout
- Fringe shifts used to determine reflectivity and phase change using 2D Fourier transform





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Spatial Interferometry

- Phase shift from change in path length of interferometer sample arm during shock

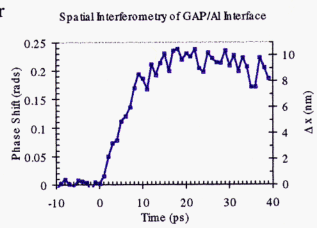
$\Delta\varphi = 2\pi\Delta(n\ d)/\lambda$

or surface motion

$\Delta x = \Delta\varphi\ \lambda/(4\pi\ n \cos\theta')$

$\theta' = \sin^{-1}((\sin\theta)/n)$

Assumes n unchanged upon shock



n is refractive index, d is distance, λ is wavelength, θ is angle of incidence

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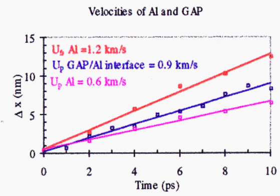
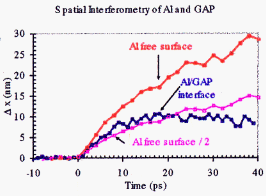
Analysis of GAP Data



•Al bulk particle velocity ~ ½ of Al free surface velocity

•Particle velocity increases entering lower density GAP



•Al/GAP interface stops moving after ~15 ps

•Rarefaction waves arriving?

•Reaction of GAP pushing back on Al surface?

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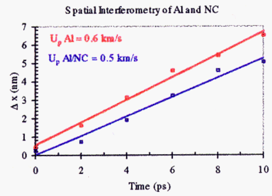
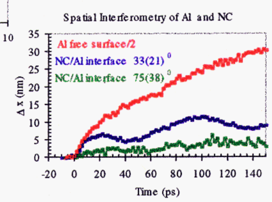
Analysis of NC Data



•U_p Al ~ U_p NC

•Oscillatory behavior at long times

•Question arises again: rarefaction or reaction?



•Effective angle in dielectric predicts 20% geometric decrease in Δx, measured 67%

•optical changes during shock disguise surface motion

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Shock Speed in NC

•Change of transit time with sample thickness directly measures shock speed

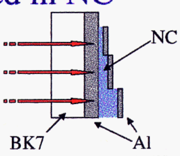
•Requires reflective overlayer (70 nm Al coated on NC)

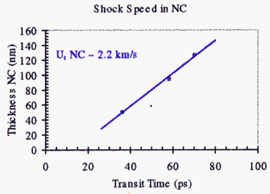
•Shock speed may decrease with thickness

•Observed U_s~ speed of sound in NC, HOWEVER,

•Large uncertainties in thickness determination

•checking with microbalance and multiple angle ellipsometry





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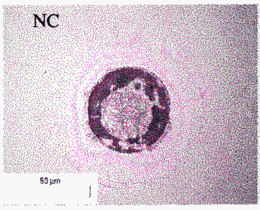
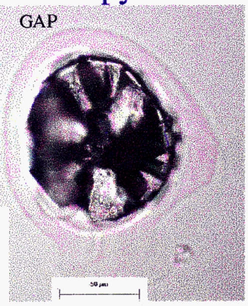
Aftershot Microscopy





•Qualitatively different

•NC has small ring with pealed Al

•GAP shows ~ 10 μm mountain of Al protruding into GAP

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Spectroscopic Probe of Reaction

•Currently looking for loss of IR absorption of GAP azide band (2100 cm^{-1}) during shock.

Possible Observations

•Simultaneous appearance of N_2 Raman band?

•Enhanced infrared absorption on red (or blue) side of peak due to population of excited vibrational states?

•Enhanced population of low frequency excited vibrations prior to high frequency excitation?

•Similar studies for NO_2 in NC, other energetics

•Other reaction products observable on time scale?

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Shocking Problems

•No Hugoniots for NC or GAP, need U_s as well as U_p

–Measure film thickness and refractive index accurately

–Solution: multiple angle and wavelength ellipsometry, double check by weighing films to within 10 μg

•Need to characterize rarefaction waves

–Solution: different Al thicknesses

•Need to isolate optical effects

•Need to provide enough sustained pressure to allow reaction

–Solution: Different drive layers

–Solution: Stretching pump pulse duration at higher energy

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Next Steps

- •More accurate knowledge of films
- •More accurate knowledge and control of shocks in the energetic polymer
- •Spectroscopically follow reaction
- •Spectroscopically follow vibrational energy transfers

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